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ENERGY DEPENDENCE OF THE TRAPPING OF URANIUM ATOMS BY ALUMINUM OXIDE SURFACES[†]

K. G. LIBBRECHT, J. E. GRIFFITH, R. A. WELLER[‡] and T. A. TOMBRELLO

W. K. Kellogg Radiation Laboratory, California Institute of Technology

Pasadena, California 91125

ABSTRACT

We have measured the energy dependence of the trapping probability for sputtered ^{235}U atoms striking an oxidized aluminum collector surface at energies between 1 eV and 184 eV. At the lowest energies approximately 10% of the uranium atoms are not trapped, while above 10 eV essentially all of them stick. In addition, we present trapping probabilities averaged over the sputtered energy distribution for uranium incident on gold and mica.

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[†] Supported in part by the National Aeronautics and Space Administration [NGR 05-002-633], the National Science Foundation [PHYS 76-83685] and the Department of Energy [EX-76-G-03-1305].

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We have measured the energy dependence of the trapping probability for sputtered ^{235}U atoms striking an oxidized aluminum collector surface at energies between 1 eV and 184 eV. At the lowest energies approximately 10% of the uranium atoms are not trapped, while above 10 eV essentially all of them stick. In addition, we present trapping probabilities averaged over the sputtered energy distribution for uranium incident on gold and mica.

1. INTRODUCTION

Most experimental studies of the trapping of slow atoms on a solid surface have involved special atom-surface combinations that allow the experimenter to overcome the formidable technical problems encountered. Consequently, data on the trapping of noble gases and alkali metals dominate the literature (Hurkmans et al.,[1,2] Sau and Merrill[3]). In this work we will present another special case, which exhibits behavior unlike any of the previously studied systems. In our experiments the incident particles were sputtered uranium atoms, which were allowed to impinge on surfaces consisting of either Al_2O_3, gold or mica. We chose uranium because there is an extraordinarily sensitive technique for detecting the fissionable isotope ^{235}U (Gregg, Switkowski and Tombrello[4]). Since the physical and chemical properties of uranium differ from those of the noble gases and the alkali metals, the technique presented here could provide important new information concerning the fundamentals of atom-surface interactions. Our own motivation for performing these experiments, however, was more practical.

During a recent series of sputtering experiments, we employed aluminum catchers to collect ^{235}U atoms (Gregg and Tombrello,[5] Weller and Tombrello,[6] and Griffith et al.[7]). To analyze these experiments we needed to know the trapping probability for the sputtered atoms striking the collector surface with the energy distribution shown in Figure 1. The collector surfaces used in these experiments were not well characterized. They were polycrystalline, oxidized and, no doubt, contaminated with a monolayer of adsorbed gas because most of our experiments were performed at 10^{-8} torr. In surface scattering experiments one usually wants to work with a scrupulously cleaned monocrystal. In our case, however, such fastidiousness was unwarranted because we were primarily interested in the efficiency of our collectors when

used under more practical conditions. Fortunately, our trapping data proved to be reproducible to within acceptable tolerances, which indicates that we had adequate control over the condition of the target surfaces.

The track detector technique employed to detect the ^{235}U atoms is not only extremely sensitive but also allows the analysis of the experiment to be simple and direct. A nuclear track detector such as mica registers fission events with unit efficiency. Thus, if a surface coated with ^{235}U is placed in contact with a mica sheet and irradiated with thermal neutrons, the tracks produced in the mica allow us to determine the uranium surface concentration. In practice, surface concentrations of 10^{10} atoms/cm^2 ($\sim 10^{-5}$ monolayers) are readily measured. This technique was employed in all of the experiments described in this paper.

Figures 2 and 3 illustrate the two types of trapping experiments that were performed. The first type (Figure 2) yields a result averaged over the energy distribution shown in Figure 1. In this experiment the sputtered atoms were produced by irradiating an isotopically enriched uranium foil with 60 keV ^+Ar. The sputtered particles were collimated but not velocity selected. The cylindrical secondary foil allowed us to determine the angular distribution of the atoms not sticking to the primary. Owing to the sensitivity of our track technique, the primary foil never accumulated more than 5×10^{13} U atoms/cm^2. The primary foil was cold rolled aluminum in most cases, but we also ran experiments with primaries consisting of mica and of gold or aluminum films evaporated onto mica. The second type of experiment (Figure 3) allowed us to determine the trapping probability as a function of energy between 1 eV and 184 eV. In it, we employed a modified version of the time-of-flight spectrometer used by Weller and Tombrello[6] to determine the energy spectrum in Figure 1. A second many-slotted wheel was added to catch uranium atoms scattering from the first

wheel. We will describe the operation of this apparatus in detail later.

2. AVERAGED TRAPPING MEASUREMENTS

The quantity we want to extract from our experiments is the fraction of incident uranium atoms not trapped by the primary collector surface. For the averaged experiment illustrated in Figure 2 we will call this fraction β. The catcher foil configuration allows us to compute β in the following manner. Suppose that N uranium atoms strike the primary foil. Of these, $N_1 = N(1 - \beta)$ are trapped while $N\beta$ are reflected. Assume that the reflected atoms strike an imaginary sphere with radius R. Of these, $N_2 = N\beta(1 - \beta')$ are trapped, where β' is the fraction not sticking to the sphere. Our data will show that β and β' are small, so to first order we have $\beta = N_2/N_1$. We are neglecting the tiny fraction $\beta\beta'$ of incident atoms that is initially trapped by neither the primary nor the secondary.

N_1 and N_2 were obtained by scanning the mica track detectors with an optical microscope. The tracks were produced by etching the micas in concentrated hydrofluoric acid after irradiating them together with the catcher foils with a known fluence of neutrons. The tracks from the primary formed a spot approximately 0.5 cm in diameter. By determining the area of the spot and the track density within it, we computed N_1. For the cylindrical secondary foil we measured the uranium surface concentration as a function of θ (see Figures 2 and 4) and then fitted the distribution to the functional form $A \cos^B\theta$ with a weighted least squares routine. This function was integrated over the imaginary hemisphere with radius R to obtain the total number of uranium atoms

$$N_2 = 2\pi R^2 \frac{A}{B + 1} .$$

In all cases the secondary was high purity aluminum foil supplied by

Ventron Alfa. This same foil was used for the primary in five runs. Because of the high purity of the foil, we did not clean it before loading it into the vacuum chamber. In one run a mica sheet (cleaved in air) was used as a primary. In two runs we employed primaries consisting of aluminum evaporated onto a mica substrate, and in two runs we used similar primaries consisting of evaporated gold. The evaporations were performed in a bell jar pumped with a liquid nitrogen chilled baffle and an oil diffusion pump. Typical pressures were about 10^{-6} torr. All of the primary surfaces were exposed to air before being loaded into the ultrahigh vacuum system. The base pressure in the UHV system was a few times 10^{-9} torr. During the runs the argon beam was the dominant gas load on the system: typical pressures were a few times 10^{-8} torr. Under these conditions we must assume that the primary and secondary surfaces were coated with at least a monolayer of contaminant gas, which could have a profound influence on the behavior of the data. Since the coverage of a surface with adsorbed gases can depend strongly on the temperature of the surface, we performed two runs at 152°C while the rest were carried out at room temperature. This allowed us to explore the stability of β against perturbations in the surface contamination. The resistance heater used to elevate the primary's temperature is illustrated in Figure 2.

The results from the averaged trapping experiments are shown in Table 1. The precision achieved in our measurement of β, the fraction not trapped, can be inferred from the scatter in the four aluminum foil runs with 80 keV argon: $\beta = 2.4\% \pm 0.3\%$. Note that we performed one run in which the uranium was sputtered with a 40 keV argon beam. A representative angular distribution is shown in Figure 4. None of the angular distributions exhibited any fine structure, so we will only quote the values of B determined from the least squares fits (Table 1).

At this point, we must consider a mechanism that could cause the results in Table 1 to be misleading. As the primary accumulated uranium atoms during a run, it was bombarded by low energy uranium atoms and higher energy argon atoms backscattered from the uranium foil. Conceivably, the uranium atoms appearing on the secondary foil were actually trapped by the primary but subsequently resputtered by the flux of atoms incident at the primary. Two simple tests were performed to explore this possibility. In the first, we performed a normal run with a ^{235}U foil to produce the sputtered uranium beam. Then we immediately repeated the run using the same catcher foils but with a ^{238}U foil to produce the sputtered beam. Track detectors are insensitive to ^{238}U, so this second run could have no influence on the final result unless the incident beam resputters ^{235}U already on the primary. The second test is conceptually similar to the first, but in this one we measured the trapping probability as a function of the ^{235}U fluence on the primary. Suppose that the incoming uranium atoms were resputtering uranium from the primary with an effective yield $\bar{S}$. The number of uranium atoms resputtered would be:

$$(\text{fluence of U atoms}) \times (\text{average surface coverage}) \times (\bar{S}).$$

The average surface coverage during the run is $\frac{1}{2}$ of the final coverage, which is proportional to the fluence of uranium atoms. Thus, the fraction not sticking to the primary would be proportional to the fluence of uranium atoms on the primary if all of the atoms reaching the secondary do so through resputtering. Similar arguments can be made if backscattered argon causes the resputtering. The results from these tests are included in Table 1. The data clearly demonstrate that β does not scale with the uranium surface density on the primary. Furthermore, the run in which the primary was also irradiated with sputtered ^{238}U did not produce an anomalous result. We may conclude that

the uranium residing on the secondary foils did not get there through resputtering.

3. ENERGY DEPENDENCE OF THE TRAPPING

We now consider the energy dependence of the fraction of incident uranium atoms not trapped, which we denote $\beta(E)$. As in the averaged experiments, we have defined

$$\beta(E) \equiv N_2(E)/N_1(E),$$

where $N_1(E)$ and $N_2(E)$ are the uranium surface densities on the primary and secondary respectively. E is the energy of the incident atom. Since we will find $\beta(E)$ to be small and slowly changing, we may write to first order

$$\beta = \int_0^\infty \beta(E)\ S(E)dE / \int_0^\infty S(E)dE$$

where S(E) is the energy spectrum of the sputtered atoms shown in Figure 1. Our measurement of $\beta(E)$ with the time-of-flight spectrometer need only be a relative measurement: we may normalize $\beta(E)$ with the data from the averaged trapping experiments.

The method by which we obtained $N_1(E)$ and $N_2(E)$ can be understood from Figure 3. The disk with only two slots corresponds to the primary foil of the previous section, while the disk with many slots is the secondary. The two disks rotate in unison at 500 Hz. Twice during each rotation the slots on the primary allow a pulse of argon atoms to pass through to the ^{235}U target, which subsequently ejects a pulse of sputtered uranium atoms with the energy spectrum S(E). When one of the slots on the secondary is aligned with the fixed slits, the atoms arriving at this disk will pass through to the primary.

Some of the uranium atoms reflected from the primary will strike the back of the secondary and be trapped. Each slot on the secondary corresponds to a given sputtered atom energy E. $N_1(E)$ and $N_2(E)$ were obtained, up to a normalization factor, by determining the fission track densities corresponding to a given slot. The track densities for $N_2(E)$ were averaged along the edges of the slots. We assumed that the angular distribution of the atoms reflected from the primary was independent of the initial energy E.

The disks were fabricated from 99.99% pure aluminum sheet, 0.051 cm thick, to minimize the background of uranium impurities. The disk radius was 5.08 cm. Into each disk we cut two diametrically opposite rectangular slots measuring 1.11 cm radially by 0.447 cm wide, which is the same size as the two fixed slits. To the secondary disk we added 28 additional slots measuring 1.0 cm radially by 0.20 cm wide, which were equally spaced at 12° intervals along the perimeter of the disk. Both disks were attached to the hub of a synchronous motor capable of rotating at 500 Hz in vacuum for extended periods. The spacing between the disks was 0.508 cm. The assembly was then dynamically balanced.

The wheel assembly was mounted in a chamber pumped with a liquid nitrogen chilled cold trap and an oil diffusion pump. During the run the pressure in the chamber was about 10^{-6} torr, which means that the wheel surfaces were contaminated with adsorbed gas. The ^{235}U target was placed 81.3 cm away from the wheel in an ultrahigh vacuum system capable of maintaining pressures of approximately 10^{-8} torr when the argon beam was on target. An in-line liquid nitrogen filled cold trap separated the two chambers. To be certain that the uranium target was clean, we sputter cleaned it before the run.

The data from this experiment are shown in Figures 5 and 6. From the paper by Weller and Tombrello[6] we know that $n_1(t)$ and $n_2(t)$ may be converted

to functions of energy through the relationship

$$E[eV] = 8.15 \times 10^5/(t[\mu\ sec])^2$$

In Figure 6 we show $\beta(E) = N_2(E)/N_1(E)$, which was normalized to give $\beta = 2.4\%$. To check the consistency of our data, the normalization factor was estimated from the wheel geometry alone. We obtained $\beta = 3\% \pm 1.5\%$, which indicates that the trapping behavior on the wheel is not grossly different from the averaged experiments.

4. DISCUSSION

The presence of unknown contaminants on the surfaces that we studied complicates any attempt at a theoretical description of the trapping. Even if the surfaces had been clean, no adequate theoretical framework exists to handle the reflection of a heavy atom from a light surface. The fact that the incoming uranium atoms are heavier than any of the atoms in the substrate means that multiple scattering must be involved when a uranium atom is backscattered. Furthermore, substrate atoms are probably displaced when the uranium atom encounters the surface, which further complicates the picture. In spite of these difficulties, some general trends and conclusions may be extracted from the data.

First of all, we note that the trapping probability is large and reproducible. Therefore, we have sufficient control over our foil surfaces to allow them to be used as catchers in sputtering experiments. In addition, the variation in $\beta(E)$ is small enough so that previous energy spectrum measurements were not seriously affected. This is in agreement with the results of Thompson et al.,[8] which showed that $\beta(E)$ was unmeasurably small ($< 10\%$) for sputtered gold incident on a contaminated steel surface. A recent series of measurements

by Weller and Tombrello[9] has shown that the trapping of Nb and Rh on Al_2O_3 surfaces behave similarly. In all of these cases the mass of the incoming atom was equal to or greater than the mass of the atoms in the substrate.

Our data differ dramatically from that of Hurkmans et al.[1,2] for sputtered alkali metals incident on clean and oxygen covered tungsten. Though they found that the presence of oxygen on the tungsten surface increased the trapping probability, it did not change the qualitative behavior of the trapping as a function of incident energy: the trapping probability decreased with increasing energy for all cases. Our data did just the reverse; the trapping increased with increasing energy. This suggests that different mechanisms are causing the trapping in the two systems. While the alkali metals are being trapped by the surface potential well, the heavy uranium atoms are most likely being trapped by imbedding themselves below the surface layer. Thus, the sharp drop in $\beta(E)$ that occurs just below 10 eV could be a manifestation of a threshold in the surface displacement mechanism.

Not all of the data fit neatly into this picture, however. Recall that two of the averaged runs were performed at a temperature of 152°C. The higher temperature should decrease the amount of contaminant gas adsorbed onto the primary surfaces, though we do not know how effective the cleaning is. The higher temperature influenced the data from both the aluminum oxide and gold films but in different ways. The trapping probability for the aluminum oxide film did not change significantly though the angular distribution of the reflected atoms became sharper at higher temperature: the exponent B in the $A \cos^B\theta$ fit increased from 0.5 to 1.1. Furthermore, the averaged trapping probability for the gold film dropped from 2.9% to 1.3% with the increase in temperature. We expected the reverse because the cleaner surface at high temperature would allow the incoming uranium atoms to interact more strongly with the heavy gold substrate, which is far more efficient than the light atoms in reflecting the uranium. Finally, the run in which we used 40 keV argon to sputter the uranium produced a somewhat lower value of β. However, since this discrepancy is of marginal statistical significance, it may not represent an additional complication in the explanation of the sticking mechanism.

These paradoxes can probably be resolved through greater control over the state of the primary surface; nothing in our technique precludes this. In fact, this method could be used with any surface not containing fissionable isotopes. The unusual results obtained so far indicate that the trapping of uranium warrants further study, and they show that this technique is a fruitful tool for exploring the behavior of surfaces bombarded with a heavy atom.

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TABLE I

Results from 10 averaged trapping probability measurements. Angular distributions of atoms scattered from the primary collector surface were fit to the form A $\cos^B\theta$.

	Primary Surface	Primary Surface Temperature (°C)	Ar Beam Energy (keV)	^{235}U Fluence on Primary ($10^{13}/cm^2$)	B	β_{tot} (%)
1.	Al Foil	23	80	4.32	.88	2.83
2.	Al Foil	23	80	1.76	.69	2.48
3.	Al Foil	23	80	2.22	.74	2.05
4.	Al Foil*	23	80	2.14	.74	2.30
5.	Al Foil	23	40	.958	.52	1.62
6.	Al Film	23	80	1.22	.52	2.55
7.	Al Film	152	80	1.28	1.05	2.17
8.	Au Film	23	80	3.60	.70	2.87
9.	Au Film	152	80	3.79	.71	1.27
10.	Mica	23	80	3.22	.56	3.02

* Also irradiated with sputtered ^{238}U.

FIGURE CAPTIONS

FIGURE 1. Energy spectrum of uranium atoms sputtered by an 80 keV $^{40}Ar^{+}$ beam as measured by Weller and Tombrello[5]. The solid line is an empirical fit to the data.

FIGURE 2. Apparatus used to measure the averaged trapping probability. The top figure shows a schematic of the arrangement used to produce the sputtered beam inside the UHV system. Each cage contained a primary-secondary assembly like the one below, though the heater was included in only two of the runs. By using two uranium foils and two shields, we could irradiate the cages separately.

FIGURE 3. The time-of-flight spectrometer used to measure the energy dependence of the trapping probability. The apparatus was located in a differentially pumped system, which kept the wheel at 10^{-6} torr and the uranium foil at 10^{-8} torr.

FIGURE 4. An angular distribution of uranium atoms scattered from an aluminum foil primary collector surface in the averaged trapping probability measurements. The oxidized foil surface was at room temperature. The uranium atoms were produced by an 80 keV $^{40}Ar^{+}$ beam. The error bars are due to statistical fluctuations in the track counts from the secondary foil. A weighted least squares fit to the data is shown; the quality of the fit and the magnitude of the exponent of the cosine distribution are typical of most of the runs.

FIGURE 5. Uranium surface densities $n_1(t)$ [primary disk] and $n_2(t)$ [secondary disk] as measured with the time-of-flight spectrometer. t is the time required

for a sputtered atom of given energy to travel from the uranium foil to the wheel. Note that the two curves are scaled differently. The error bars were calculated from statistical fluctuations in the counted track densities. The curve $n_1(t)$ was taken from Figure 1.

FIGURE 6. Energy dependence of the trapping probability for uranium atoms incident on an aluminum oxide collector surface as inferred from the data in Figure 5 and Table 1.

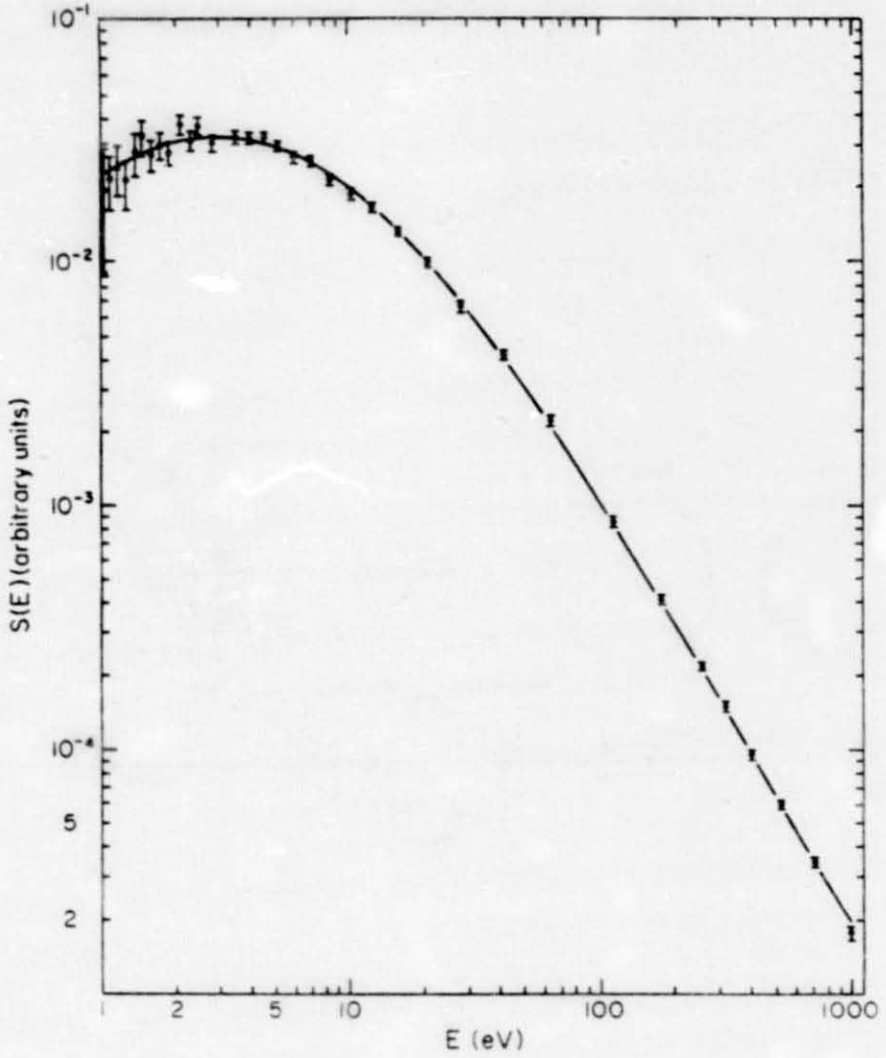

FIGURE 1

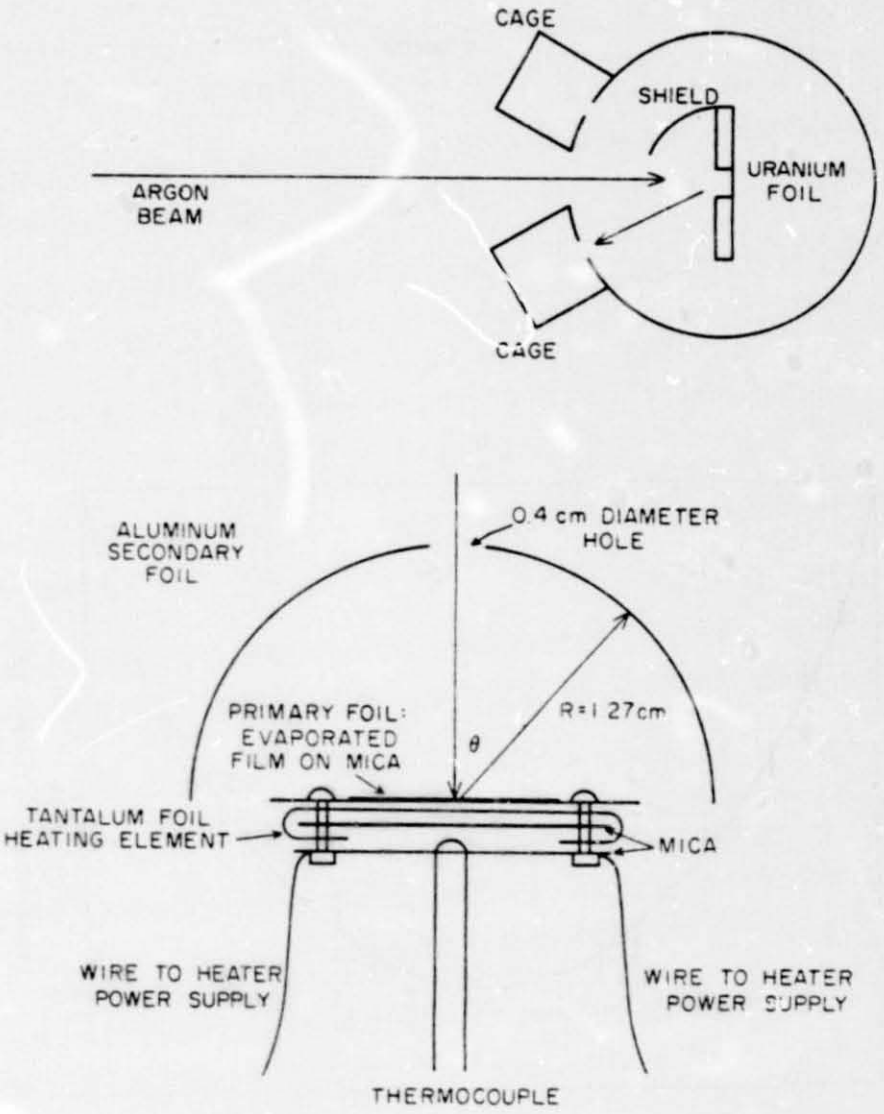

FIGURE 2

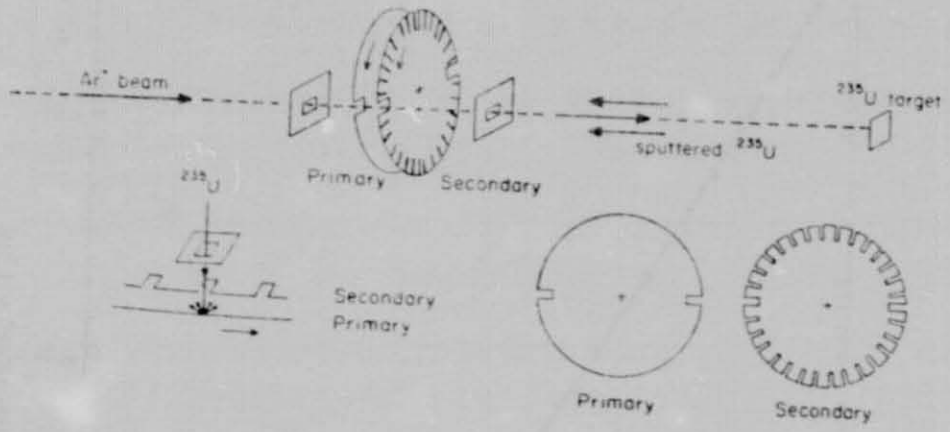

FIGURE 3

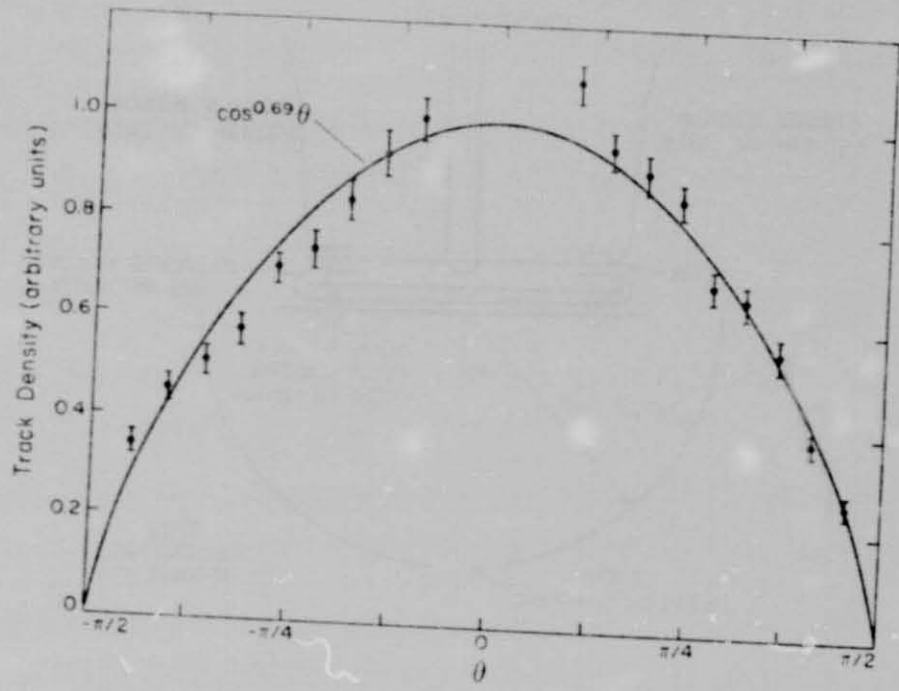

FIGURE 4

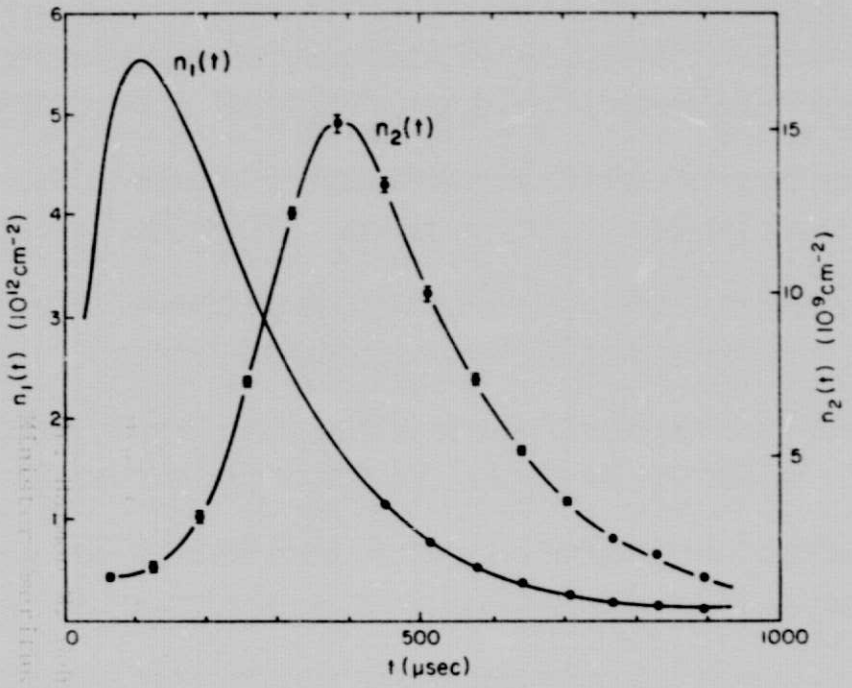

FIGURE 5

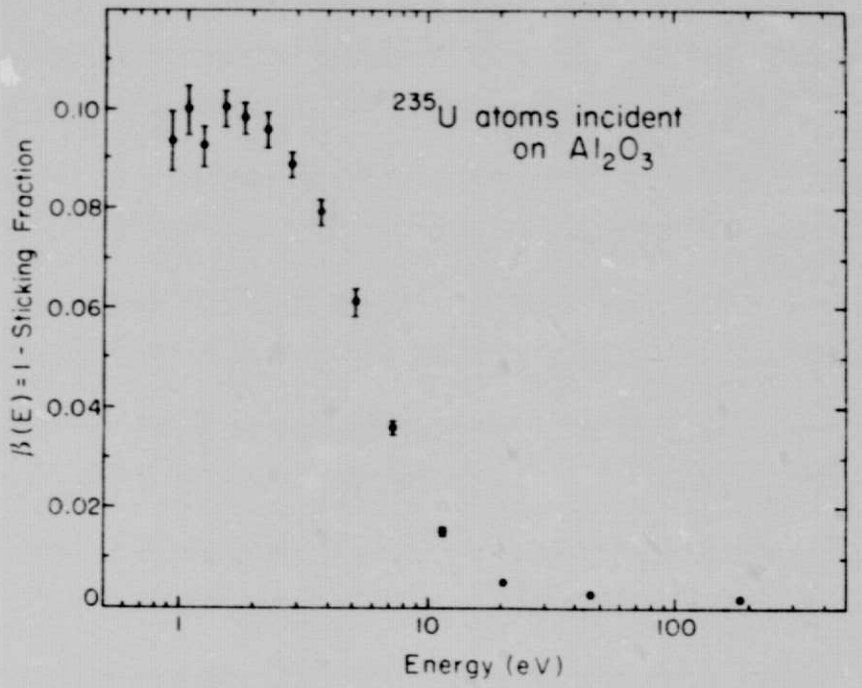

FIGURE 6